GEOPHYSICAL RESEARCH LETTERS, VOL. 27, NO. 23, PAGES 3917-3920, DECEMBER 1, 2000

Radon-222 Monitoring of Soil Diffusivity

Bernhard E.Lehmann and Martin Lehmann
Physics Institute, University of Bern, Switzerland

Albrecht Neftel
Institute for Environmental Protection and Agriculture (IUL), Bern-Liebefeld, Switzerland

Sergei V.Tarakanov
Institute of Silicate Chemistry, St.Petersburg, Russia

Abstract. The temporal evolution of activity profiles of the radon isotope ^{222}Rn in the top 50 cm of soil is used to quantify diffusive gas transport in the air-filled pore space. Air is continuously collected from gas-permeable, hydrophobic membrane tubes placed at various depths under the surface and circulated through a high-sensitivity Rn detector. With a micro-processor controlled inlet system an automatic monitoring of several levels is possible for extended periods of time with a typical time resolution of one hour. In addition, a new dynamic approach to measure the in-situ soil diffusivity D_s (m^2/s) around the subsurface tubes is presented where the recovery from air injection is evaluated every 3 hours. Within a test period of 3 weeks D_s dropped from 10^{-6} m^2/s to $3\,10^{-7}$ m^2/s at a depth of z = 50 cm due to increasing water content of the soil.

1. Introduction

Radon-222 is a radioactive noble gas isotope with a half-life of $T_{1/2}$ = 3.82 days. Atoms are continuously generated in rocks and soils in the natural α-decay series of ^{238}U; the mother radionuclide of ^{222}Rn is ^{226}Ra. A fraction of the Rn atoms emanates from the site of production into the air-filled pore space of e.g. soil, from where some reach the surface by diffusion and escape to the atmosphere.

A knowledge of the gas transport characteristics of soil is not only essential to estimate the public health hazard of Rn, it is also of general importance for understanding soil-plant interactions and trace gas exchange between soil and atmosphere. Changes in trace gas concentrations (e.g. CO_2, CH_4, N_2O, NO) in soil can be caused by physical as well as by numerous chemical and/or biological processes and it is usually difficult to separate them in a complex system. A measurement of Rn concentrations in parallel to the measurements of reactive species simplifies this task since physical factors only need to be considered for radon. Once the transport properties of the soil are quantified it is easier to understand concentration variations of reactive trace gases.

Radon isotopes have been used in a number of environmental studies (*Wilkening, 1990*) and variations of the activity in soil gas were investigated (*Gates et al., 1990; Washington and Rose, 1992, Nazaroff 1992*) . Usually researchers had to collect individual soil gas samples for these analyses. Here we present a new analytical approach for the continuous monitoring of Rn concentrations in soil gas at various depths below the surface with high temporal resolution (typically one hour) which enables a much more detailed study of the transport processes in the air-filled pore space of soils.

2. Analytical Technique

New commercial radon gas monitors (ALPHAGUARD, Genitron Instruments, Frankfurt, Germany) have been combined with a microprocessor controlled gas sampling system for the continuous monitoring of ^{222}Rn- and ^{220}Rn-activities (Bq/m^3) above and below the surface [*Lehmann et al.*, 1999]. By applying digital signal processing to pulses from an ionization chamber a detection limit of about 3 Bq/m^3 for a data accumulation time of 10 minutes is achieved (corresponding to about 1 Rn decay in the detector volume of 0.63 litres). A small membrane pump transports air first through a delay volume where ^{220}Rn atoms (half-life 56 seconds) decay and then through an aerosol filter to remove radon daughter products before entering the detector. For

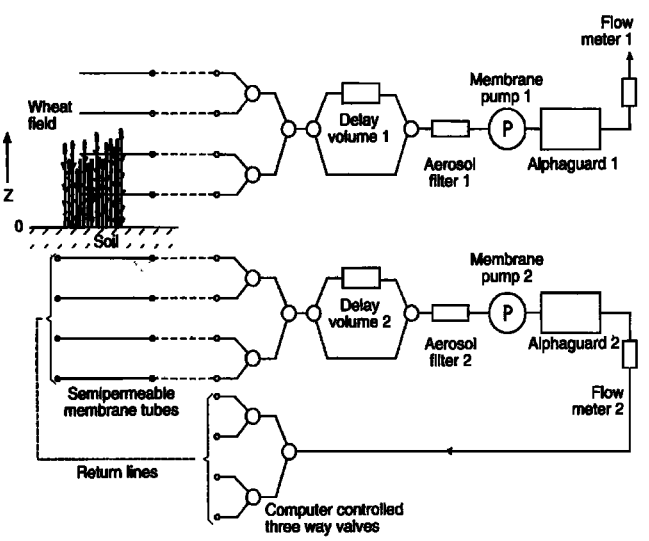

Figure 1 : Experimental set-up for continuous measurements of ^{222}Rn- and ^{220}Rn-activities above and below the surface.

Paper number 1999GL008469.
0094-8276/00/1999GL008469$05.00

^{220}Rn-analysis the delay volume is bypassed. Programmable valves switch between different inlet lines.

To collect air from below the surface we have developed a membrane tube technique (METT) (*Gut et al.*, 1998) which is based on gas diffusing from the air filled pore space into circulating air in an air-permeable, hydrophobic, polypropylene tube. For the data presented below sections of 2.5 m length were placed at various depths into an agricultural grass field prior to the growing season. The tubes have an inner diameter of 5.5 mm and a mean wall thickness of 1.5 mm. In laboratory tests it was demonstrated that the maximum volumetric air flow inside the membrane tube is 1.2 liters/minute to reach a permeation efficiency close to unity in this situation. The Rn activity at the outlet of the membrane tube is then equal to the activity in the soil gas outside the tube.

For a total volume of 1340 cm^3 (620 cm^3 delay volume, 620 cm^3 detector volume, 100 cm^3 tubing) and a flow rate of 0.16 liters/minute the circulation time (one round-trip) is 8.4 minutes. In such a situation the air transit time through the 0.62 liter delay volume is close to 4 minutes and therefore 95% of the ^{220}Rn atoms decay before reaching the detector. The remaining activity is due to ^{222}Rn. Data acquisition can be set to 1 min, 10 min or 1 hour intervals.

3. Time Series of ^{222}Rn-Activity

As an example a continuous record of ^{222}Rn-activities at four different depths below the surface is presented in Figure 2. The data were taken at our experimental field site Kerzersmoos which is located 20 km north-west of Bern in a flat rural area of the Swiss plateau. The time series covers six consecutive days without precipitation. During this time the soil started to dry out causing a sequential change of the Rn activities at the selected depths. Near the surface (z = 10 cm) the water filled porosity dropped from 45% to 30%. The activities at 5 cm and at 10 cm drop continuously; at 25 cm the decrease only starts on the 3rd day while at 50 cm it stays essentially constant over this time period.

For each measuring interval of 60 minutes at one depth the detector measured six activities (Bq/m^3), one every 10 minutes. The data of the first two of these intervals were ignored because after switching from one z-level to the next a certain time is necessary to bring the circulating gas back to equilibrium with the soil gas at the new depth. The other four data points were averaged to get a mean concentration at the selected depth during the time interval of one hour.

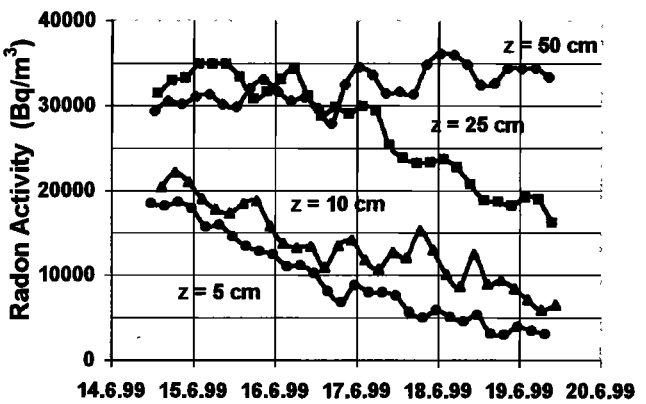

Figure 2 : Radon activity at 4 depths during 6 days.

4. Vertical ^{222}Rn-Activity Profiles in Soil Gas

The one-dimensional diffusion equation (1) for radioactive atoms has to be solved, where I_a is the radon activity per volume of soil gas (Bq/m^3). Table 1 gives the meaning of all the other parameters following the nomenclature given in the excellent review of radon transport from soil to air (*Nazaroff.,1992*), as well as numerical values which are representative for an average situation in our experimental soil.

$$(\varepsilon_a + \kappa\varepsilon_w + \rho K)\partial_t I_a = \varepsilon_a D_e \partial_z^2 I_a - \lambda I_a(\varepsilon_a + \kappa\varepsilon_w + \rho K) + fA_{Ra}\lambda\rho \quad (1)$$

The effective diffusivity D_e (m^2/s) is smaller than the diffusivity D_o in air due to the tortuous nature of the pores and the fact that (in particular in wet soils) some of the possible pathways between two points are blocked by water.

Dividing (1) by the factor $(\varepsilon_a + \kappa\varepsilon_w + \rho K)$

gives the simpler form

$$\partial_t I_a = D_s \partial_z^2 I_a - \lambda I_a + P \quad (2)$$

where the soil diffusivity D_s takes into account the Rn fractions in air, dissolved in water and sorbed on soil grains. (For the numerical values given in Table 1 the respective fractions are 57%, 37% and 6%). It is related to the effective diffusivity D_e by

$$D_s = \frac{\varepsilon_a D_e}{\varepsilon_a + \varepsilon_w \kappa_w + \rho K} \quad (3)$$

P is the production rate of "mobile" Rn per volume of soil gas given by

$$P = \frac{\lambda f \rho A_{Ra}}{\varepsilon_a + \varepsilon_w \kappa_w + \rho K} \quad (4)$$

In a homogeneous medium and with all parameters being constant in time the solution of (2) is mathematically simple but may not be very realistic. It will, however, give a first overall range of numerical values for the Rn-activities (Bq/m^3) one might expect in soil gas. Integration yields a steady-state exponential activity profile below the surface according to

$$I_a(z) = I_\infty\left(1 - e^{-z/z^*}\right) \quad (5)$$

The scale depth z* which is a characteristic length over which a Rn atom travels before it decays is related to the soil diffusivity D_s by

$$z^* = \sqrt{\frac{D_s}{\lambda}} \quad (6)$$

The maximum activity at great depth follows from

$$I_a(z\to\infty)=\frac{P}{\lambda}=\frac{f\rho A_{Ra}}{\varepsilon_a+\varepsilon_w\kappa_w+\rho K}=I_\infty \quad (7)$$

and the diffusive flux J^d (per unit area of bulk porous medium) at depth z can be calculated from

$$J^d(z)=-(\varepsilon_a+\varepsilon_w\kappa_w+\rho K)D_s\partial_z(I_a(z)) \quad (8)$$

At the surface (flux from soil to atmosphere) one gets

$$J^d(0)=-(\varepsilon_a+\varepsilon_w\kappa_w+\rho K)D_s\frac{I_\infty}{z^*}=-\lambda f\rho A_{Ra}z^* \quad (9)$$

Table 1 : Average numerical values for the investigated soil

radium activity of soil [1]	A_{Ra}	40 Bq/kg
emanation coefficient from soil [2]	f	0.25
air-filled soil porosity [2]	ε_a	0.17
water-filled soil porosity [2]	ε_w	0.38
total soil porosity (= $\varepsilon_a+\varepsilon_w$) [1]	ε	0.55
solids density [1]	ρ_g	2700 kg/m^3
bulk density of soil (=$\rho_g(1-\varepsilon)$) [3]	ρ	1215 kg/m^3
aqueous-gaseous partition coeff. [1]	κ_w	0.29 (at 15 °C)
sorbed-gaseous partition coeff. [2]	K	$1.4\,10^{-5}$ m^3/kg
Rn decay constant [1]	λ	$2.1\,10^{-6}$ s^{-1}
diffusivity of Rn in air [1]	D_o	$1.2\,10^{-5}$ m^2/s
effective diffusivity [3]	D_e	$2.6\,10^{-7}$ m^2/s
soil diffusivity [2]	D_s	$1.5\,10^{-7}$ m^2/s
production rate of mobile Rn [3]	P	0.086 Bq/(m^3 s)
equilibrium activity ($z\to\infty$) [3]	I_∞	40900 Bq/m^3
scale depth [3]	z*	0.27 m
diffusive Rn-flux to the atmosphere [3]	J^d	0.0068 Bq/(m^2.s)

[1] known or measured parameter, [2] representative average value
[3] calculated

The numerical values of Table 1 are used to calculate the depth profile in Figure 3 as a comparison to five experimental profiles taken at midnight, respectively, from the data of Figure 2. Obviously, the soil conditions change faster than the characteristic time $\tau = 1/\lambda = 5.5$ days in which the Rn-concentrations would approach a steady-steady exponential

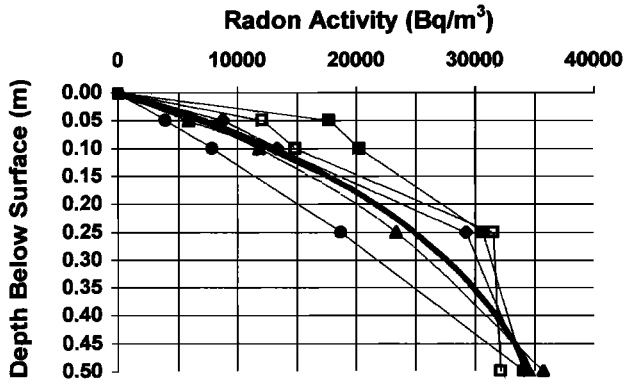

Figure 3 : Five experimental Rn depth profiles (at midnight each from the data in Figure 2) compared to an average exponential profile calculated from the parameters of Table 1.

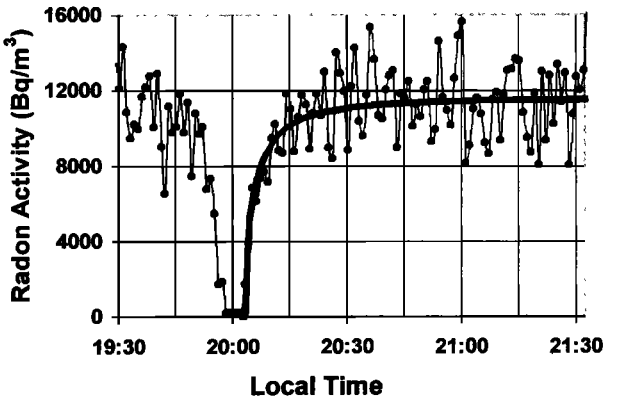

Figure 4. Raw data of the Rn-detector operated in the one-minute-data acquisition mode. Prior to the a cycle of 3 hours the detector is flushed with Rn-free air. Solid line : calculated recovery curve for $D_s = 7.10^{-7}$ m^2/s.

profile. Furthermore, as the soil is first drying out near the surface there must be a continuous change of the diffusivity at each depth with time. The calculated curve (from the values of Table 1 with $D_s = 1.5\,10^{-7}$ m^2/s) clearly only approximates the real situation. Therefore, a more detailed investigation of the temporal evolution of the soil diffusivity at a selected depth is necessary.

5. Recovery from Air Injection : A Dynamic Measurement of the Soil Diffusivity

In Figure 4 the raw data from our radon detector are presented for a time period of 2 hours when data were accumulated every minute. At 19:50 the volumes of detector and ^{220}Rn-delay line were flushed for 10 minutes with atmospheric air which is essentially Rn-free. This removed a well defined amount of Rn from the circulating air. After switching back to the underground membrane tube radon is diffusing back from a cylindrical volume of soil air around the tube into the circulating air.

Within about 20 minutes the activity is back at the old level prior to the flushing; the recovery time being determined by the local diffusivity around the membrane tube. This output activity at the end of the tube as a function of time has been calculated by solving numerically the diffusion equation in cylindrical symmetry. An iterative procedure was used to determine the best fit to measured recovery curves such as the one presented in Figure 4.

A simple calculation can illustrate the geometric relations. With a detector volume and a ^{220}Rn delay volume of 620 cm^3 each, the Rn removed from the system during the flushing interval corresponds to the Rn content in a hollow cylinder with a radius of only 2.3 cm around the membrane tube (tube length 2.5 m, outer diameter of membrane tube 8.5 mm, air and water filled porosities from Table 1). Taking this radius as a first approximation for the mean distance the replacement radon has to diffuse during the recovery time τ of approx. 20 minutes, the soil diffusivity can immediately be estimated from $D_s = r^2/\tau = 4.4\,10^{-7}$ m^2/s.

The results of an extended field test are presented in Figure 5. The system continuously operated for three weeks at the

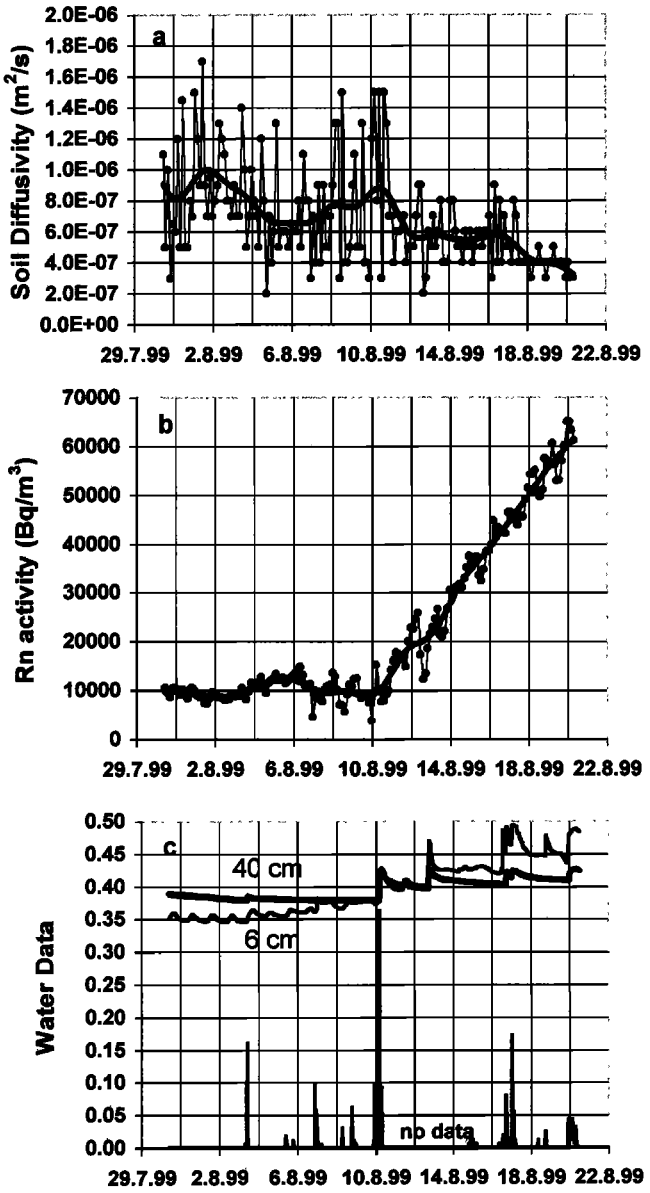

Figure 5 : (a) Soil Diffusivity D_s at z = 50 cm, (b) Radon activity at z = 50 cm, (c) Water data. Top : Water-filled soil porosity at z = 6 cm (thin curve) and at z = 40 cm (thick curve) and precipitation (bottom) (multiply by 30 to get mm of rain per 30-minute accumulation interval).

selected depth of z = 50 cm with data acquisition running at one- minute intervals. Every three hours the flushing procedure described above was automatically repeated yielding a total of 168 individual in-situ measurements of the soil diffusivity D_s. Its value decreased by about a factor of three (smoothed curve in plot 5a) during the 3 weeks.

During this time the Rn activity at the same depth (Figure 5b) in soil air increased from 10 to 60 kBq/m^3. Heavy rain on August 10 and additional precipitation in the second half of the observation period considerably changed the volumetric soil moisture (= water-filled soil porosity in Table 1). Note that on Aug 12 rain was not recorded due to technical problems with the rain gauge; the volumetric soil moisture , however, clearly shows an increase at both depths of 6 cm (top of Figure 5c, thin curve) and of 40 cm (top of Figure 5c, thick curve).

Obviously the diffusive transport of Rn towards the surface was slower in the second have of the period giving rise to a dramatic increase of the activity at 60 cm depth in spite of only a rather moderate decrease of the diffusivity at this depth. It appears that the very wet top layers of the soil almost completely blocked gas transport from below. Additional experiments and a more detailed numerical model of the air, water and radon budgets in soil are in preparation. The presented first results of our new analytical technique demonstrates the potential for such future work.

6. Discussion and Outlook

Diffusion in the air-filled pore space of soils is slower than in air because of the tortuous nature of the pores and the fact that water can block some of the possible pathways between two points. It is however not simple to quantify the relation between the diffusivity of a gas in air and that in a particular soil for a given water content of soil. With the dynamic approach presented above it is possible to directly measure the soil diffusivity D_s as defined by (3) of Rn and correlate it to soil properties (water-filled pore volume, field capacity, pore geometry). Such studies are in progress.

The use of radon as inert tracer in soil physics was demonstrated with our experimental set-up in two recent studies a) to calibrate the NO flux from the soil to the atmosphere [*Gut et al.*, 1999] and b) to help establish the scale length of N_2O in the soil of a grassland [*Neftel et al.*, 2000].

Acknowledgements. This project was supported by the Swiss National Science Foundation and the University of Bern. It is part of a Swiss collaboration within BIATEX-2 (Biosphere-Atmosphere-Exchange) of the European Environmental Research Program EUROTRAC-II. The technical help of H.P.Moret and H.Riesen is greatly appreciated.

References

Gates A.E., L.C.S.Gundersen, L.D.Malizzi. Comparison of Radon in Soil over Faulted Crystalline Terranes: Glaciated versus Unglaciated, *Geophysical Research Letters*, 17, 6, 813-816, 1990.

Gut, A., A.Blatter, M.Fahrni, B.E.Lehmann, A.Neftel, T.Staffelbach. A new membrane tube technique (METT) for continuous gas measurements in soils, *Plant and Soil 198*, 79-88, 1998.

Gut, A., A.Neftel, T.Staffelbach, M.Riedo, and B.E.Lehmann. Nitric oxide flux from soil during the growing season of wheat by continuous measurements of the NO soil-atmosphere concentration gradient : A process study, *Plant and Soil 216*, 165-180, 1999.

Lehmann B.E., M.Lehmann, A.Neftel, A.Gut, S.V.Tarakanov. Radon-220 Calibration of Near-Surface Turbulent Gas Transport, *Geophysical Research Letters*, 26, 5, 607-610, 1999.

Nazaroff W.W. Radon Transport from Soil to Air, *Reviews of Geophysics*, 30, 2, 137-160, 1992.

Neftel A., A.Blatter, M.Schmid, B.E.Lehmann, S.V.Tarakanov. An Experimental Determination of the Scale Length of N_2O in the Soil of a Grassland, *Journal of Geophysical Research*, 105, D10, 12095-12103, 2000.

Washington J.W., A.W.Rose. Temporal Variability of Radon Concentration in the Interstitial Gas of Soil in Pennsylvania, *Journal of Geophysical Research*, 97, B6, 9145-9159, 1992.

Wilkening M. Radon in the Environment, in *Studies in Environmental Sciences 40*, Elsevier, 137 pages, 1990.

(Received October 15, 1999; revised July 7, 2000; accepted September 13, 2000.)